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TITLE: Nanostructures: Physics and Technology. 7th International Symposium. St. Petersburg, Russia, June 14-18, 1999 Proceedings

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7th Int. Symp. "Nanostructures: Physics and Technology" St Petersburg, Russia, June 14–18, 1999 © 1999 Ioffe Institute

LO phonon mediated relaxation in InP self assembled quantum dots in electric field

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Abstract. Strong LO phonon resonances are found in the photoluminescence spectra of InP self-assembled quantum dots under an applied bias. These arise from fast phonon assisted relaxation when the applied bias suppresses the photoluminescence.

Introduction

In the optical spectra of semiconductor quantum dot structures with not very high quantum yield of the photoluminescence (PL), features due to optical phonon mediated relaxation of excitation are observed $[\ \]$. In high quality structures, on the other hand, when the main recombination channel for excited electron hole pairs is radiative, there are no such features. In present work we found that applying an electric field to InP QDs leads to decreasing of the quantum yield. As a result, features caused by LO phonons become observable in the PL spectra.

1 Experimental result

The studied heterostructure QDP1779 was grown by gas source molecular beam epitaxy on an n⁺ GaAs substrate. The QDs were formed by the deposition of InP on the InGaP layer and covered by the top InGaP layer. The areal density of the QDs is about 10^{10} cm⁻². Average base diameter is ≈ 50 nm and height is ≈ 10 nm. The sample was provided with a semi-transparent gold Shottky contact on the top surface and an ohmic contact on the back surface.

The PL spectra were recorded by using a cw Ti:sapphire laser, a double monochromator U1000 and a photon counting system with cooled GaAs photomultiplier tube. All the measurements were done at 5 K.

The PL spectra of the InP QDs under negative bias ("-" is on the top of the sample) are shown in Fig. 1(a). Without bias there are no strong features in the spectrum. When a negative bias is applied the intensity of the PL decreases and distinct resonances appear in the spectrum. As the negative bias is increased, these resonances grow in intensity relative to the rest of the PL band. However, under rather strong bias ($U_{\rm bias} < -1.0 \, \rm V$) intensities of the resonances start to decrease. Energy gaps between the laser line and the resonances approximately correspond to the 1LO and 2LO phonon energies in InP. As is seen from Fig. 1(b), the 1LO resonance has a rather complicated structure. A decomposition of this structure into Gaussian functions give the phonon energies 41.0 meV, 43.7 meV, 45.3 meV, 47.2 meV and 48.0 meV. The spectral shape of the 2LO resonance is reproduced by the convolution of the 1LO resonance.

QWR/QD.06 25

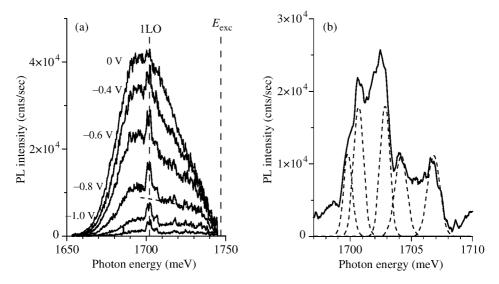


Fig. 1. (a) Dependence of the PL spectrum on the applied electric field. The excitation energy is 1747 meV. The positions of the excitation photon energy and the 1LO resonances are marked by the vertical dashed lines. (b) Part of the PL spectra around the 1LO resonance ($E_{\rm exc} = 1747$ meV, $U_{\rm bias} = -0.8$ V). A decomposition of the resonance into Gaussians is shown by the dotted lines. The signal was accumulated by the detection of the PL from the large illuminates sample area of about 0.03 mm². A background signal is substrated according the dashed line shown in the Fig. 1(a).

In Fig. 2(a) the dependence of the QDs PL spectra on the excitation photon energy $E_{\rm exc}$ is shown. The energy positions of the 1LO and 2LO resonances follow $E_{\rm exc}$. Phonon energies determined by the Gaussian decomposition of the resonances do not depend on $E_{\rm exc}$. The intensity of the resonances depends on their position with respect to the PL band. The integral intensity of the 1LO resonance has a spectral dependence that coincides very well with the PL spectrum (Fig. 2(b)). The 2LO resonance becomes weak when it goes out of the PL band.

2 Discussion

The presented experimental data strongly suggest that the observed resonances are caused by LO phonon mediated relaxation processes rather than by phonon sidebands of the resonant PL or by resonant Raman scattering. An applied bias leads to the suppression of the PL from the QDs due to the activation of various nonradiative processes.

The rates of these processes become comparable or higher than the phonon relaxation rates of the hot carriers under the increase of negative bias. It is known that a relaxation involving the LO phonons is faster than the acoustic phonons [2]. Therefore the PL in the region of the LO resonance is not suppressed so strong as in the region formed by the acoustic phonon relaxation.

For semiquantitative analysis we consider a simple model illustrated in Fig. 2. Due to small depth of the hole potential well in QDs [3] an applied bias activates a tunneling of the holes from the QDs into barrier layer. A tunneling rate exponentially depends on bias U in low rate limit: $\gamma = \gamma(\infty) \cdot e^{-U_0/U}$ [4].

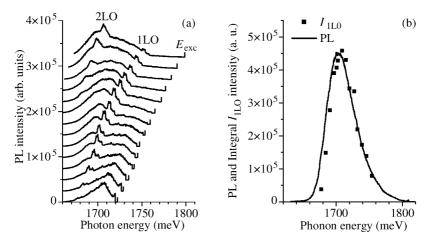


Fig. 2. (a) Dependence of the PL spectra on the excitation energy $E_{\rm exc}$. $U_{\rm bias} = -0.8$ V. (b) The spectral dependences of the QDs PL under high $E_{\rm exc}$ and zero bias (solid line) and the integral intensity $I_{\rm LO}$ of the 1LO resonance (solid squares).

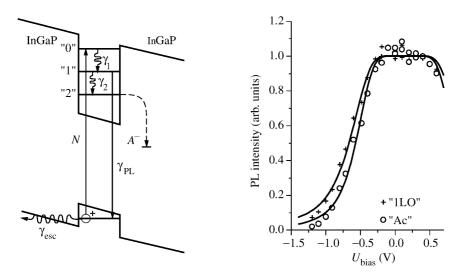


Fig. 3. A simplified model of the PL suppression in electric field.

Fig. 4. The dependence of the normalized PL intensity I_{PL}^* on U_{bias} in the "1LO" and "Ac" spectral regions fitted by the expression (1).

An analysis shows that the PL suppression can be modelled as

$$I_{\rm PL}(U) = \frac{I_{\rm PL}(0)}{(1 + a \cdot e^{-U_0/U}) \cdot (1 + b \cdot e^{-U_0/U})}.$$
 (1)

Here a, b and U_0 are the fitting parameters. As an example, in Fig. 2 a normalized PL intensity $I_{\text{PL}}^* = \frac{I_{\text{PL}}(E,U)}{I_{\text{PL}}(E,0)}$ in dependence on U_{bias} is presented for two neighboring spectral

QWR/QD.06 27

ranges $\Delta E = 35-38$ meV ("Ac" range) and $\Delta E = 44-47$ meV ("1LO" range) under the excitation photon energy $E_{\rm exc} = 1747$ meV.

It is seen that the fitting reproduces well the general behavior of PL and its difference for "Ac" and "1LO" spectral ranges in spite of many simplifications used in the model. A good agreement between the experimental data and a fitting is observed also for the spectra recorded at the other $E_{\rm exc}$.

A discovered phenomenon presents an interesting possibility to study the different resonances in the PL spectra. As is seen in Fig. 1(b), a PL spectrum in the region of the LO resonance is formed by the several peaks. According the data [\degree , \degree], two lowest peaks with the energy distance from $E_{\rm exc}$, $\Delta E=41.0$ meV and $\Delta E=43.7$ meV are maybe caused by the LO phonons of the InP QDs in Γ and $\bf X$ (or $\bf L$) points of Brillouin zone. The former peaks at 45.3 meV, 47.2 meV and 48.0 meV are caused by the LO phonons of the InGaP surrounding material.

In the frame of the proposed attribution, an observation of the intensive peaks in PL spectra caused by the LO phonons of the InGaP layers is evidence a strong penetration of the electron (and maybe hole) wave functions into barrier layer.

3 Conclusion

Our experimental data show that the observation of LO resonances is in PL is possible if the quantum yield is not high. It is a good test of the optical quality of the structure. The study of the resonance structure gives useful information about the phonons in InP QDs and the surrounding material.

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